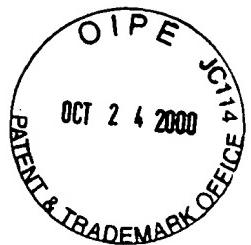


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GP1764/3
10/24/00PATENT
Attorney Docket No. 10151-1IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re Patent Application of:

)

W. Roth et al.

)

Serial No. 09/305,019

)

Examiner: Tam Nguyen

Filed: May 4, 1999

)

Group Art Unit: 1764

APPELLANTS' BRIEF UNDER 37 CFR 1.192(a)

Fairfax, Virginia 22030

Commissioner for Patents
Washington, D.C. 20231

Sir:

Appellants appeal to the honorable Board of Patent Appeals and Interferences the Primary Examiner's final rejection of the claims set forth in the Office Action of Examiner Nguyen mailed July 20, 2000.

As required, the Appeal Brief is being filed in triplicate and is accompanied by a check in the amount of \$310 under 37 CFR 1.17(c).

1. Status of the Claims

Claims 4-10, inclusive, are before the Board for consideration.

2. Status of Amendments

Appellants have not filed any amendments after final rejection.

3. Summary of the Invention

The present invention relates to an alkylation/transalkylation process for preparing a monoalkylated compound from alkylatable aromatic compound and alkylating agent.

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Liquid phase alkylation/transalkylation processes are of increasing commercial importance since, by operating at lower temperatures, they result in lower yields of unwanted by-products. However, such liquid phase processes suffer from the problem that their lower operating temperatures increase the activity requirements of the catalyst particularly in the transalkylation step. According to the invention, it has now been found that small crystal (<0.5 micron), TEA-mordenite has unexpectedly high activity when used as a liquid phase transalkylation catalyst in a separate transalkylation reactor. In particular, small crystal TEA-mordenite exhibits unexpectedly higher activity as a liquid phase transalkylation catalyst than conventional TEA-mordenite as shown by Example 4 of the present application.

4. Issues

- a. Did the Examiner err in rejecting Claims 4-9 under 35 U.S.C. § 103(a) as being unpatentable over EP 0733608 ('608) in view of U.S. Patent No. 5,557,024 to Cheng et al. (Cheng)?
- b. Did the Examiner err in rejecting Claim 10 under 35 U.S.C. § 103(a) as being unpatentable over EP 0733608 ('608) in view of U.S. Patent No. 5,557,024 to Cheng et al. (Cheng) and further in view of U.S. Patent No. 4,891,458 to Innes et al. (Innes)?

5. Grouping of Claims

Appellants submit that process claim 10 which recites specific transalkylation conditions which are neither suggested nor disclosed by the '608 or Cheng references stands separately from the remaining claims which are not so limited

6. Arguments

A. Claims 1-9 have been finally rejected under 35 U.S.C. § 103(a) as being unpatentable over EP 0733608 (the '608 reference) in view of U.S. Patent No. 5,557,024 (Cheng et al.).

The Examiner argues that the '608 reference discloses a transalkylation process of polyalkylbenzenes which are produced *during* alkylation of benzene with olefins, e.g.,

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ethylene, by using a crystal aluminosilicate catalyst having an average size of less than 0.5 micron. The alkylation and transalkylation processes (which appear to occur simultaneously within the same reactor) are conducted at a temperature between 250°C and 500°C, a pressure between 200 psi and 500 psi (1390-34447 kPa), a WHSV of 20 to 150 hr⁻¹, and a feeding ratio of benzene to ethylene of 2:1 to 20:1. The reference is also cited as disclosing that a transalkylation process can be operated in a separate reactor. (However, there is no actual teaching of using '608's small crystal size aluminosilicate catalyst as a transalkylation catalyst in a separate transalkylation reactor—only as an alkylation catalyst having transalkylation activity, see, e.g., '608 reference, page 2, lines 25-28).

The Examiner admits that '608 fails to specifically disclose 1) that the alkylation/transalkylation step is operated in the liquid phase, 2) MCM-56 as alkylation catalyst and 3) TEA-mordenite transalkylation catalyst. Accordingly, he cites Cheng as disclosing alkylation and transalkylation processes that use MCM-56 alkylation catalyst and TEA-mordenite transalkylation catalyst. The Examiner concludes it would have been obvious to one of ordinary skill in the art at the time the invention was made to modify the '608 process by using MCM-56 as an alkylation catalyst and TEA-mordenite as a transalkylation catalyst as taught by Cheng which also discloses that alkylation and transalkylation processes can be conducted in either vapor phase or liquid phase. Appellants respectfully disagree with the Examiner's conclusion and request the Board to consider the following arguments.

The '608 reference differs from the present invention in several important ways.

First, the reference relates to an aromatics alkylation/transalkylation process which is conducted in the vapor phase (see page 2, line 19 and the conditions disclosed on page 3, lines 20 and 44) while the present invention requires liquid phase transalkylation.

Secondly, the '608 reference's polyalkylated species is recycled to the alkylation reactor (see claim 1, lines 2 and 3) in contrast to the present invention's transalkylation zone which is separate from the alkylation reactor. Although the Examiner refers to the

statement on page 2, line 21 that the polyalkylated species in conventional alkylation/transalkylation processes can be fed to a separate transalkylation reactor, it is clear from the ensuing disclosure at page 2, lines 21-31 that the invention in the '608 reference is a process in which the polyalkylated species is recycled to the alkylation reactor for transalkylation. Thus, according to page 2, lines 29-31, the discovery of the '608 reference is that the transalkylation activity of the *alkylation catalyst* (emphasis added) is enhanced if the alkylation catalyst has a crystal size of less than 0.5 micron. Accordingly, one skilled in the art acquainted with the '608 reference would not likely substitute its combination alkylation/transalkylation catalyst for the single purpose transalkylation catalyst which is employed in the present invention.

Thirdly, even assuming the '608 catalyst were considered by one skilled in the art as substitutable in the present invention's alkylation step, the reference is still of little use to one seeking a transalkylation catalyst for a separate transalkylation step. Apart from small crystal size, the only guidance the '608 reference gives as to the type of alkylation catalyst which exhibits increased transalkylation activity is that the catalyst should be an aluminosilicate with a Si/Al atomic ratio of 50 to 500 (see claim 1) and the aluminosilicate should have a primarily monoclinic structure (see page 3, line 21). No importance is attached in the '608 reference to the type of zeolite used as the alkylation/transalkylation catalyst and certainly there is no disclosure or suggestion in the reference of the use of small crystal TEA-mordenite as a *separate* transalkylation catalyst

Indeed, the specific low crystal size catalyst as taught by the '608 reference actually teaches *away* from the TEA-mordenite zeolite of the present invention in several respects! While the former is a primarily *monoclinic* structure which may not contain more than 40% orthorhombic crystalline structure (see page 3, lines 19-25), it is well known that mordenite exhibits an orthorhombic crystalline structure (*see, e.g., APPENDIX A, Meir and Olson, "Atlas of Zeolite Structure Types," Butterworth-Heinemann, 1992, p.144*). Moreover, the '608 reference specifically teaches that "the pore size of the catalyst is also noteworthy, ranging from about 1000Å to about 1800Å" (page 3, line 36). Such large pore size clearly militates against substitution by the present invention's relatively small (7Å) pore size TEA-mordenite!

Cheng is cited by the Examiner to overcome the '608 reference's admitted shortcomings in i) failing to disclose that the alkylation/transalkylation step is operated in the liquid phase, ii) failing to disclose MCM-56 alkylation catalyst and TEA-mordenite transalkylation catalyst, and iii) failing to disclose operating the transalkylation in a separate reactor. The Examiner characterizes Cheng as disclosing alkylation and transalkylation processes wherein MCM-56 or zeolite beta is used in an alkylation zone, while TEA mordenite is used to transalkylate alkylation by-products such as polyalkylbenzene. The Examiner concludes it would have been obvious for one skilled in the art at the time the invention was made to have modified the '608 process by operating in the liquid phase because Cheng teaches operating alkylation and transalkylation in either vapor or liquid phase, and transalkylation in a separate reactor. The results of such modification would be expected to be similar or the same as the present invention, according to the Examiner.

Relying on 35 USC 103(a)'s standard "that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains," it is reasonable to refer to the Examiner's own cited prior art (including the '608 reference) for determining the state of the art in which the present invention was made. In particular, it is reasonable to assert that one skilled in the art at the time the invention was made would look to the references relied upon by the Examiner, including the teachings of the '608 reference regarding characteristics of a suitable catalyst. Despite Cheng's teaching of TEA-mordenite in transalkylation processes, it is respectfully submitted that one skilled in the art acquainted with the '608 reference's disclosure of using small crystal size aluminosilicate in transalkylation, would not combine the two references. First of all, the aluminosilicate of the '608 reference is described as having pore sizes 140 to 260 times greater than Cheng's TEA-mordenite! One skilled in the art would be led away from the present invention inasmuch as it would not be reasonable to assume one could substitute TEA-mordenite for 1000-1800 angstrom pore size material without radically affecting the products produced.

In making this argument, appellants urge that the Examiner should have considered the reference as a whole and was not free to discard portions of a reference which are less than accommodating to the position he wishes to take. See, *In re Young*, 18 USPQ2d 1089, 1091 (Fed. Cir. 1991) wherein it was stated "patents are part of the literature of the art and are relevant for *all* they contain (emphasis added)." See, also, *Gillette Co. v. S.C. Johnson & Son, Inc.*, 16 USPQ2d 1923, 1927 (Fed. Cir. 1990) wherein it was stated that the closest prior art reference "would likely discourage the art worker from attempting the substitution suggested by (the inventor/patentee)."

Assuming, *arguendo*, that it is fair to substitute a small crystal TEA-mordenite for the small crystal catalyst in the '608 reference, the resulting process would be directed to a method wherein the transalkylation catalyst is employed in an *alkylation* reactor using *recycled* polyalkylaromatics, rather than the present invention's use of a separate transalkylation reactor. Moreover, while Cheng teaches the use of TEA-mordenite as a separate transalkylation catalyst, Cheng's TEA-mordenite is synthesized according to the disclosure of U.S. Patent Nos. 3,766,093 and 3,894,104. As shown by Examples 2 and 3 of the present application, TEA-mordenite produced according to these earlier patents has a crystal size of *>5 micron!* Indeed, inasmuch as the '608 reference specifically discloses small crystal size catalyst in a single alkylation/transalkylation step, one skilled in the art combining these references would not substitute '608's small crystal catalyst for the TEA-mordenite of Cheng's separate transalkylation reactor, but rather for the MCM-56 in Cheng's *alkylation* reactor. Finally, Cheng is silent as to the phase conditions of the transalkylation step, particularly where the transalkylation is performed in a separate reactor from the alkylation step.

Accordingly, it is respectfully submitted that this combination of references fails to disclose or suggest in any way the present invention. In view of this, reversal of the Examiner's final rejection by the Board of claims 4-9 is respectfully requested.

B. In the final rejection of claim 10, U.S. Patent No. 4,891,458 to Innes et al. (Innes) is additionally cited as specifying operating conditions. In particular, Innes

discloses transalkylation at 50-100 psig with benzene/polyalkylated benzene weight ratios of 1:1 to 50:1. Reversal of this rejection by the Board is respectfully requested in view of the following arguments.

Despite Innes' teachings, it is respectfully urged that its combination with the '608 reference and Cheng fails to disclose or suggest the present invention, in view of the arguments made above respecting the latter two references.

Innes teaches a process for producing alkylaromatic compounds in which an aromatic compound is alkylated with a C₂-C₄ olefin in the presence of zeolite beta in an alkylation zone under at least partial liquid phase conditions. After separation of the aromatic feed and the desired monoalkylated product, the polyalkylated fraction is contacted with additional aromatic compound in the presence of zeolite beta in a transalkylation zone under at least partial liquid phase conditions. Even assuming for argument's sake that it would be obvious to combine and modify the processes of the '608 reference and Cheng so as to adopt the teaching of Innes, the present invention is neither disclosed nor suggested because Cheng suggests the use of a zeolite beta as a transalkylation catalyst. It is respectfully submitted that such a combination would therefore employ *zeolite beta* rather than *TEA-mordenite* in the transalkylation reactor. It would be directly contrary to the teaching of Innes to effect a combination in which small crystal TEA-mordenite is employed as the transalkylation catalyst. Thus the subject matter of claim 10 is patentably distinguished from the combination of Innes, the '608 patent and Cheng.

Accordingly, it is respectfully submitted that this combination of references fails to disclose or suggest in any way the present invention. In view of this, reversal of the Examiner's final rejection by the Board of claim 10 is respectfully requested.

7. Conclusion

Appellants respectfully submit that the foregoing arguments obviate all of the Examiner's final outstanding rejections in this case. The cited references neither disclose nor suggest the improved transalkylation activity of small crystal size TEA-mordenite in a liquid phase transalkylation process. In view of this, reversal of these rejections by the Board is respectfully requested.

Respectfully submitted,

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October 23, 2000

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APPENDIX

CLAIMS:

4. A process for producing a monoalkylated aromatic compound comprising the steps of:
 - (a) contacting an alkylatable aromatic compound with an alkylating agent in the presence of an alkylation catalyst in an alkylation reactor to provide a product comprising said monoalkylated aromatic compound and a polyalkylated aromatic compound, and then
 - (b) contacting the polyalkylated aromatic compound from step (a) with said alkylatable aromatic compound in the liquid phase and in the presence of a transalkylation catalyst in a transalkylation reactor separate from said alkylation reactor, said transalkylation catalyst comprising TEA-mordenite having an average crystal size of less than 0.5 micron to produce said monoalkylated aromatic compound.
5. The process of claim 4, wherein the alkylation step (a) is conducted in the liquid phase.
6. The process of claim 4, wherein the alkylating agent includes an alkylating aliphatic group having 1 to 5 carbon atoms.
7. The process of claim 4, wherein the alkylating agent is ethylene or propylene and the alkylatable aromatic compound is benzene.
8. The process of claim 4, wherein the alkylation catalyst of step (a) is selected from MCM-22, MCM-49, MCM-56 and zeolite beta.

9. The process of claim 4, wherein step (a) is conducted at a temperature between about 300° and 600°F (about 150° and 316°C), a pressure up to about 3000 psig (20875 kPa), a space velocity between about 0.1 and 20 WHSV, based on the ethylene feed, and a ratio of the benzene to the ethylene between about 1:1 and 30:1 molar.

10. The process of claim 4, wherein step (b) is conducted at a temperature of 100 to 260°C, a pressure of 10 to 50 barg, a weight hourly space velocity of 1 to 10 on total feed, and benzene/polyalkylated benzene weight ration 1:1 to 6:1.

TABLE OF CASES

1. In re Young, 18 USPQ2d 1089, 1091 (Fed. Cir. 1991).
2. Gillette Co. v. S.C. Johnson & Son, Inc., 16 USPQ2d 1923, 1927 (Fed. Cir. 1990).

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on October 23, 2000.

A handwritten signature in black ink, appearing to read "L P Hobbes". It consists of a stylized "L" followed by a more fluid "P Hobbes".

Laurence P. Hobbes

APPENDIX A

ATLAS OF ZEOLITE STRUCTURE TYPES

W.M. Meier and D.H. Olson



Third Revised Edition
1992

Published on behalf of the
Structure Commission of the International Zeolite Association

by

Butterworth-Heinemann

London Boston Singapore Sydney Toronto Wellington

MOR

MORDENITE

Cmcm

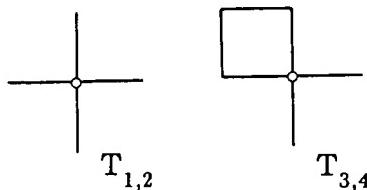
Secondary building units:

5-1

Framework density:

17.2 T/1000 Å³

Loop configuration of T-atoms:



Coordination sequences:

$T_1(16)$ 4 12 22 38 60 88 115 155 204 242
 $T_2(16)$ 4 12 20 37 64 87 114 154 198 241
 $T_3(8)$ 4 11 24 39 54 86 126 156 195 242
 $T_4(8)$ 4 11 24 39 60 92 122 148 195 250

Channels:

[001] **12** 6.5 x 7.0* ↔ [010] **8** 2.6 x 5.7*

Fault planes:

(010), (100)

Type material:

Mordenite $\text{Na}_8[\text{Al}_8\text{Si}_{40}\text{O}_{96}] \cdot 24 \text{ H}_2\text{O}$
orthorhombic, Cmcm, $a=18.1$, $b=20.5$, $c=7.5$ Å⁽¹⁾

Isotypic framework structures:

Na-D⁽²⁾ Large port mordenite⁽⁴⁾
Ca-Q⁽³⁾ LZ-211⁽⁵⁾
Zeolon

Alternate designations:

Ptilolite Flokite (discredited)
Arduinite (discredited)

References:

- (1) W. M. Meier, Z. Kristallogr. **115**, 439 (1961).
- (2) R. M. Barrer and E. A. D. White, J. Chem. Soc. **1952**, 1561 (1952).
- (3) M. Koizumi and R. Roy, J. Geol. **68**, 41 (1960).
- (4) L. B. Sand, Molecular Sieves, Soc. of Chem. Industry, London, p. 71 (1968).
- (5) D. W. Breck and G. W. Skeels, US Patent 4,503,023 (1985).